Isotopic studies of the Oklo fossil reactors and the feasibility of geological nuclear waste disposal

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Abstract

In 1972, six 2000 million year old natural fossil nuclear fission reactors were discovered at a uranium orebody at Oklo in Gabon, South West Africa. Over a period of about one million years the reactors produced some hundreds of tonnes of fission products with characteristic but unnatural isotopic compositions. These isotopic compositions have enabled the nuclear characteristics of the reactors and the extent to which they have retained their fission products to be determined. Since their discovery another dozen fossil fission reactors have been found in Gabon. This paper summarizes the results obtained from both the earlier and recent discoveries and their implications for the storage of anthropogenically generated fission waste in geological environments.

Introduction:

The existence of natural fossil chain fission reactors is perhaps one of the most unusual discoveries in the comparatively short history of nuclear science. When Fermi and colleagues initiated the first artificial self sustaining chain fission reactions they did not realize that nature had assembled and operated a similar type of reactor 2000 million years before. Fourteen years after Fermi's achievements, Kuroda (1956) published the details of the conditions under which such reactors could exist. However, despite an extensive survey of the $^{235}U/^{238}U$ ratio during the 1960s, no variation greater than analytical uncertainty (±<0.1%) was found. In 1972, routine measurements of $^{235}U/^{238}U$ in a French uranium processing plant revealed a depletion in this ratio of 0.43%. At first it was thought that this was caused by "spent fuel" from a conventional fission reactor but it was eventually traced to uranium from a sedimentary type uranium deposit at Oklo in Gabon, South West Africa. A survey of the existing Oklo mine site revealed six metre long and tens of centimetres thick lenses of highly enriched U which contained both depleted $^{235}U/^{238}U$ ratios and substantial quantities of nuclear fission products. The circumstance giving rise to these fossil nuclear reactors has subsequently become known as the "Oklo Phenomenon".

The potential significance of the Oklo fossil reactors as natural analogs for geological repositories of radioactive waste was quickly recognized by the scientific community. Although the possibility of geological storage had been considered prior to the discovery of the Oklo reactors, the methods of assessing waste retention over the time required for it to decay to "safe" levels were extremely limited. The long term behaviour of such waste had to be extrapolated from extremely short term experiments under conditions which poorly approximated those likely to be found in real repositories. Direct experimentation of sites through trial deposition of hundred of tonnes of fission waste was also potentially environmentally irresponsible. Besides, the waste problem needs to be tackled now and cannot wait for the time required to assess the potential of geological storage.

de Laeter Symposium on Isotope Science
Curtin University of Technology, Perth, 1995

The role of isotopic studies:

Isotopic studies have proved to be the master key for unlocking the many secrets of the Oklo phenomenon. Although non-isotopic studies of the Oklo reactors may reveal unusual amounts of some elements it is only by measuring their isotopic composition that the existence of nuclear fission can be confirmed. Isotopic studies by mass spectrometry are especially powerful because they allow the fission products to be distinguished from the natural elements and their amounts measured in any rock sample.

The wealth of information available from isotope studies can be illustrated using tellurium as an example. Figure 1 shows the relative isotopic compositions of five of the major isotopes of tellurium as measured in a range of samples. For comparison all isotope abundances are referenced to the ^{130}Te abundance taken as unity. The solar system values shown are those present in all terrestrial sources, meteorites and the moon (Smith et al., 1978). Samples ORZ9-028 and ORZ9-006 represent Te from samples from inside reactor zone 9 while OHR9-002 is from a host rock sample taken 1.4 m away from this reactor zone.

The isotopic abundances of ^{128}Te and ^{130}Te in ORZ-028 are essentially the same as that produced by the fission of ^{235}U. Since no ^{124}Te is produced by fission, the absence of this isotope indicates that no terrestrial Te is present in the reactors and that all of the Te present in the reactors is fissiogenic.The ^{128}Te abundance in OHR9-002 indicates that this sample contains a mixture of both fissiogenic and solar system Te. The other isotope of significance is ^{126}Te which in excess in samples ORZ9-006 and in particular OHR9-002. This effect arises from the parent isotope, ^{126}Sn which has a half life 10^5 years indicating that radioactive tin is mobilized inside and in the immediate

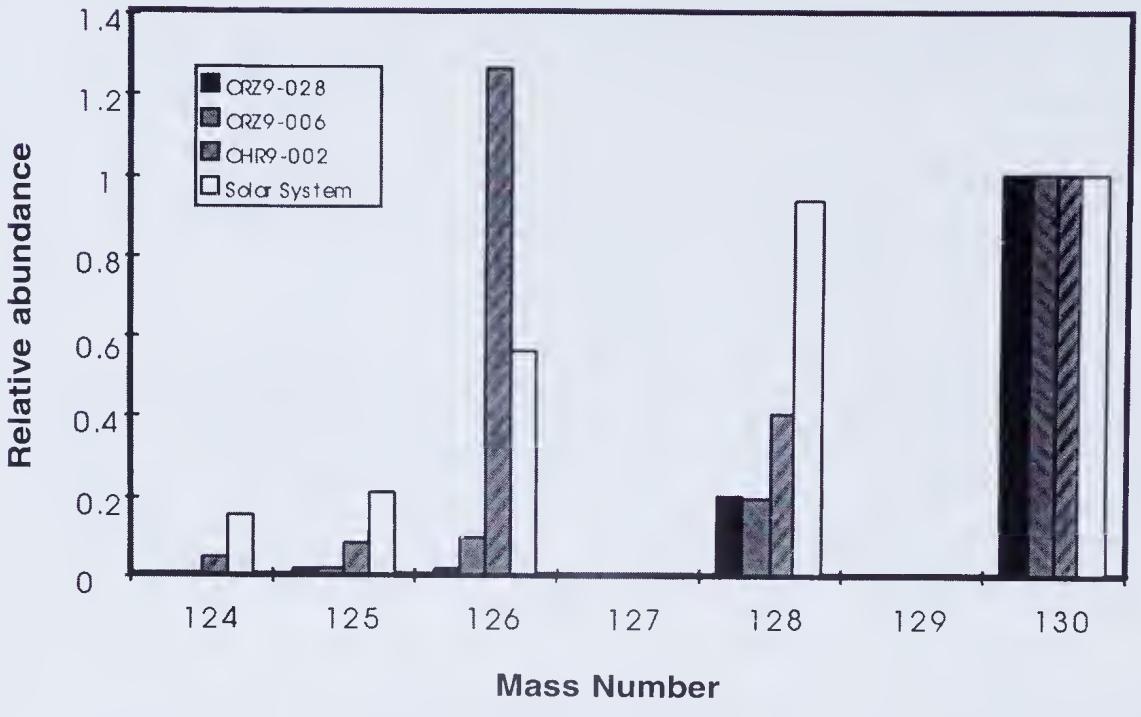

Figure 1. The isotopic composition of tellurium in a range of Oklo and other samples (data from Curtis *et al.* 1989 and Loss *et al.* 1989)

vicinity of the reactors during the reactions and within one million years (10 half lives) of the last reaction. The abundance of fission product Te present in OHR9-002 was also measured to be 3 orders of magnitude lower than that in the reactor zone while at a distance of six metres from the reactor zone the abundance of fission product Te is 1 part in 10^5 of that inside the reactors.

Besides confirming the occurrence of the fission process at Oklo, the isotopic compositions of the fission products enabled the nuclear characteristics of the reactors themselves to be determined (Naudet 1975 and Ruffenach et al., 1980). The parameters of interest include the integrated neutron flux (fluence), the degree of thermalization of the neutron spectrum, and the fractional fission contribution from ^{238}U and ^{239}Pu. From these parameters, other physical characteristics such as the power output and temperature of the reactors and surrounding host rocks can be determined. These parameters, together with the age of the reactors, the degree of U enrichment and other geochemical parameters have been used to calculate the total quantity of fission products expected to be produced by the reactors. When this is compared to what is present today a retention factor for the fission products can be determined. For example, reactor zone 9 has retained more than 80% of its fissiogenic Te - a remarkable achievement for a 2000 million year old uranium oxide mineral assemblage located inside a highly porous sandstone matrix.

Isotopic studies have shown that many fission produced elements have been retained inside the reactor zones (Curtis et al., 1989). The rocks surrounding the reactors have also retained other fission products within a few metres of the reactor zones (Loss et al., 1989). These results are in excellent agreement with those predicted by thermodynamics and geochemistry (Brookins, 1975). However, there are still many aspects of the reactors that remain to be studied.

Recent discoveries

Since 1972, another dozen or so fossil fission reactors have been discovered at Oklo, Oklobondo and at Bangombe, some 30 km further south of Oklo. These reactors are significant discoveries in their own right since they include a wider range of geological conditions than those represented by the original reactors. One of these reactors is located within metres of a 1000 million year old doleritic dike which cut through the centre of the Oklo uranium ore deposit. This has provided the opportunity to study the affect of dike emplacement on the retention of fission products within a reactor. Although only a limited number of studies of these newer reactors have been undertaken the retention of fission products appears to be similar to those for the original reactors. Amongst the most significant new results has been the detection of previously unstudied but very important long lived radiogenic and highly mobile nuclei, ^{137}Cs (decaying to ^{137}Ba) and ^{90}Sr (decaying to ^{90}Zr) within metres of reactor zones. These elements were previously thought to have been completely lost from the vicinity of the reactors (Hidaka et al., 1993 and 1994). Another unusual discovery has been the formation of unusual refractory and highly insoluble alloys which have trapped almost pure fissiogenic elements within the original uraninite ore grains (Gauthier-Lafaye et al 1996). Studies of this type of alloy may prove useful in developing a suitable storage matrix for some types of spent fuel.

Perhaps the most unusual of the new reactors are those at Bangombe which are much closer to the surface and considerably more weathered and oxidized than those at either Oklo or Oklobondo. Studies of this type of reactor will be particularly useful since future underground waste sites may eventually be exposed to surface conditions and more aggressive geochemical environments. Isotopic studies of minerals within and surrounding this reactor zone has identified clays and phosphates

as important in retaining rare earth element fission products. A case in point is the detection plutonium retention identified by a higher than solar system $^{235}U/^{238}U$ ratio (Bros *et al.* 1993). This has occurred in what has essentially been a high rainfall equatorial environment for perhaps millions of years.

Conclusions

The most important lesson that Oklo teaches us is that nature is able to isolate fissiogenic waste in geological environments for extremely long periods of time even though the reactors were contained in a highly fractured, porous and physically "leaky" matrix. This geophysical environment is one that would hardly be considered as ideal for a nuclear waste repository. The reason for the retention of fissiogenic materials under these conditions was largely due to the local geochemical conditions. The oxidizing potential and acidity of the ground waters together with the presence of scavenging clays and other minerals played a critical role in this process. However it can be concluded that the concept of multiple barrier geological containment including iron oxides and clays which are able to buffer possible changes in ground water conditions indicates that the effective containment of man-made nuclear waste is feasible.

References

Brookins D G 1977 Applications of the Eh-pH diagrams to problems of retention and/or migration of fissiogenic elements at Oklo. International Symposium of the Oklo Phenomenon, Libreville, Gabon. IAEA TC-119/33:243-265.

Bros R, Turpin L, Gauthier-Lafaye F, Holliger P & Stille P 1993 Occurrence of naturally enriched ^{235}U: Implications for plutonium behaviour in natural environments. Geochemie et Cosmochimie Acta 57:1351-1356.

Curtis D B, Benjamin T M, Gancarz A L, Loss R D, Rosman K J R, de Laeter J R, Delmore J E, & Maeck W J 1989. Fission product retention in the Oklo natural reactors. Applied Geochemistry 4:49-62.

Gauthier-Lafaye F, Holliger P & Blanc P-L 1996 Natural fission reactors in the Franceville Basin, Gabon: A review of the conditions and results of a "critical event" in a geological system. Geochim et Cosmochim Acta: in press.

Hidaka H, Holliger P & Masuda A 1993 Evidence of fissiogenic Cs estimated from isotopic deviations in a Oklo natural Reactor zone. Earth and Planetary Science Letters, 114:391-396.

Hidaka H, Sugiyama K, Ebihara M. & Hollinger P. 1994 Isotopic evidence for the retention of 90Sr inferred from the excess of 90Zr in the Oklo natural fission reactors. Earth and Planetary Science Letters, 122:173 -182.

Kuroda P 1956 On the nuclear Stability of Uranium Minerals. Journal Chemical Physics 25:781 -782.

Loss R D , Rosman K J R, de Laeter J R, Curtis D B, Benjamin T M, Gancarz A L, Maeck W J & Delmore J E 1989 Fission product retentivity in peripheral rocks at the Oklo natural fission reactors, Gabon. Chemical Geology 76:71-84.

Naudet R. 1975 Mechanismes de regulation des reactions nucleaires. In: The Oklo Phenomenon. Symposium Proceedings, Libreville, IAEA, Vienna 589-600.

Ruffenach J C, Hagemann R & Roth E 1980 Isotopic Abundances measurements - a key to understanding the Oklo Phenomenon. Zeitschrift fuer Naturforsch ung 35A:171-179.

Smith C L Rosman K J R & de Laeter J R 1978 The isotopic composition of tellurium. International Journal of Mass Spectromatry and Ion Physics 28:7-17.